

WASTE MINIMIZATION IN THE LOS ALAMOS MEDICAL RADIOISOTOPE PROGRAM

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ABSTRACT

Since the mid-1970s the Los Alamos Medical Radioisotope Program has been irradiating target materials to produce and recover radioisotopes for applications in medicine, environmental science, biology, physics, materials research, and other disciplines where radiotracers find utility. By necessity, the chemical processing of targets and the isolation of radioisotopes generates radioactive waste materials. There have been in recent years federal mandates requiring us to discontinue the use of hazardous materials and to minimize radioactive waste volumes. As a result substantial waste reduction measures have been introduced at the irradiation facility, in processing approaches, and even in the ways the product isotopes are supplied to users.

INTRODUCTION

For two decades the Medical Radioisotope Program at The Los Alamos National Laboratory has been recovering radioisotopes from targets irradiated at the Los Alamos Meson Physics Facility (LAMPF).¹ The 800 MeV proton beam with currents up to 1 milliAmpere provides unique irradiation conditions for radioisotope production. With these high currents and energies come many waste handling challenges. The targeting

equipment at LAMPF is subjected to extreme conditions resulting in the need to periodically replace and dispose of highly radioactive components. Spallation reactions result in a large number of isotopes associated with a diverse group of elements produced in each target.² The chemical recovery of individual radioisotopes requires specific and sometimes elaborate procedures resulting in the production of radioactive waste materials in the form of contaminated reagents and labware. The use of returnable lead shields and packing for shipping radioisotopes results in contaminated lead shields and packaging material that must be cleaned or discarded on their return.

The laboratory has a radioactive waste disposal site which makes it possible to dispose of radioactive waste provided it is not contaminated with hazardous materials. Hazardous materials include carcinogens, toxic metals, flammable solvents, etc. The laboratory does not have a facility to dispose of hazardous materials but relies on an outside contractor for this service. Mixed waste is a combination of radioactive and hazardous materials. At this time, neither the laboratory nor outside contractors have a mechanism to dispose of this type of material. In recent years we have been subject to more restrictive regulatory, financial, and moral directives to eliminate mixed waste and minimize hazardous waste materials. Consequently, as we have developed approaches and techniques to perform irradiations, process targets, and provide radioisotopes to users, we have also pursued ways to manage the unique waste generated by this work.

METHODS

LAMPF Irradiation Facility

The LAMPF irradiation facility employs 9, twenty-six foot long target stringers.³ Each stringer supports a target in the proton beam of the accelerator and provides cooling water to the target. Originally the core of each stringer was filled with concrete containing lead shot for radiation shielding. Over time, the program has had to replace

some of these stringers due to mechanical failures caused by the extreme operating environment. Because lead is a hazardous material these stringers were considered a form of mixed waste. The lead shot in the core has since been replaced with magnetite, making them no longer a mixed waste when replacement is required. Engineering changes including bearings, welded water line fittings, and alignment fixtures have extended the life of new stringers thus reducing the contribution to the total volume of radioactive waste. In addition, we now remove and reuse the portion of the stringer that is not highly activated thus reducing the associated waste even more.

Video cameras are used to monitor remote loading and removal of targets at the irradiation facility. In the past, these cameras have required replacement at frequent intervals due to failure caused by the high radiation environment. Shielded enclosures with leaded glass windows have greatly extended the life of these cameras and protected them from contamination and activation thus reducing this contribution to the radioactive waste stream.

Modifications to the remote handling tools used for the loading and removal of targets have made it possible to replace damaged portions of the tooling without replacing the entire tool. These modifications also insure that if a tool is broken, the failure is not in the remote environment and the damaged portion is easy to replace.

The external target encapsulation, called a carrier, is made of aluminum and was initially bolted closed.⁴ The seals for this closure could not tolerate long irradiations, and thus carriers for such irradiations required frequent replacement. As a result of extensive testing of various aluminum alloys and closure mechanisms, we now use forged 2219 aluminum alloy target encapsulation that is welded closed. This arrangement precludes replacing the carrier during long irradiations. Minimizing the size of the carrier has also reduced both the bulk and the amount of radioactivity associated with the carrier when it is discarded. The approach of replacing only the damaged portion of a piece of equipment, coupled with materials and engineering improvements has made a substantial reduction in the waste associated with the irradiation of targets.

Elimination of Mixed Waste from Target Processing

A major program objective has been the elimination of mixed waste produced in target processing. Because spallation produces a variety of elements and radioisotopes in the targets, our recovery procedures require a diversity of chemical separation techniques. Unfortunately many traditional and some new chemical separation approaches employ hazardous materials. For example, solvent extraction procedures have been a common mechanism for performing radiochemical separations. Unfortunately many extractants, or the solvents used with them, have been found to be hazardous materials. Ion exchange procedures have often been enhanced by the incorporation of water miscible organic compounds. Many of these compounds are now considered hazardous. Gaseous isotope recovery procedures often have employed hazardous materials to make cold temperature baths. Many reagent gases used in chemical procedures are now considered hazardous. Disposal of cylinders of Partially used hazardous gases associated with radioactive contamination has become extremely difficult.

The elimination of hazardous materials associated with the recovery of radioisotopes has posed significant challenges but not without some unexpected benefits. For example, early procedures for recovering aluminum-26 from potassium chloride targets employed an extraction step using acetylacetone.⁵ This hazardous compound is no longer used; instead a precipitation step is employed to separate the aluminum isotope from the target solution, followed by dissolution and classical ion exchange methods. This new procedure provides an extremely valuable radiotracer with no mixed waste generation and reduced waste volume as compared to the previous method.⁶ Acetylacetone was also used in an extraction step to isolate vanadium-48,49 and beryllium-7 from irradiated zinc oxide targets.⁷ An alternative procedure has been developed that employs a precipitation step followed by conventional ion exchange techniques to isolate these isotopes.

A good example of our efforts to eliminate mixed waste is the processing of molybdenum targets. Molybdenum targets are processed principally for the recovery of Sr-82. Five or six of these targets are processed each year. Each target produces two to twelve curies of Sr-82 depending on the length of irradiation. For many years we used an ion exchange procedure that employed as much as 4.5 liters of dioxane for each target.⁸ Elimination of the dioxane from the chemistry was accomplished by dissolving the molybdenum metal in hydrogen peroxide instead of a mixture of nitric and phosphoric acid.⁹ This change eliminated the mixed waste problem but the volume of waste increased from an average of 11.5 liters to 17.2 liters. This change also resulted in a more difficult target dissolution and precipitation problems. Additional work resolved the precipitation problems making it possible to decrease molybdenum solution volumes. Eliminating the phosphoric acid from the procedure also made it possible to recover zirconium-88, a feature not possible using the original procedure.¹⁰ Finally, a third procedure was developed that utilized the improvements in the hydrogen peroxide dissolution, and further reduced waste volumes.¹¹ The final procedure also increased the Sr-82 yield and had the added benefit of making it possible to recover technetium, rubidium, selenium, arsenic, and germanium isotopes.^{12,13} The flow charts demonstrate the evolution of our Sr-82 recovery procedures.(Figures 1,2,3)

Minimization of Target Processing Waste Volume

Target processing waste is primarily composed of hot cell fixtures, plasticware, glassware and reagents. Corrosion is the biggest contributor to the failure and subsequent disposal of hot cell hardware. Plasticware and glassware are used to introduce reagents to the hot cells and to perform chemical separations. The chemical processing of targets results in the distribution of different radionuclides in the used reagents. The laboratory does not have a system for treating a liquid waste stream containing high levels of the isotopes produced in our targets. These reagents must be neutralized and converted to a

solid form before disposal which increases the volume by two to four times the original volume. Therefore it is important to minimize liquid waste volumes.

The program's efforts to minimize the replacement of hot cell fixtures has focused on reducing chemical corrosion in the cells and a modular approach to equipment used. The use of spill trays, good housekeeping, and condensers during evaporation procedures are the most effective approaches in controlling chemical corrosion. Taking these steps has greatly extended the life of hot plates, pH meters, radiation detector probes, and other electrical appliances and has reduced the frequency at which hot cells require refurbishment. This represents a substantial reduction in radioactive waste, since refurbishment requires disposal of the entire contents of the cell, decontamination, and replacement of all appliances and hardware. Using equipment with replaceable, modular parts in a remote environment minimizes down time and further reduces the volume of the waste stream.

Plasticware and glassware is saved and reused for the processing of additional targets and often for the subsequent recovery procedures of other product isotopes from the same targets. Bulk associated with plasticware is primarily in the form of plastic bottles; many of these are reused as primary containers for neutralized and sorbed reagent waste. Used glassware is crushed to minimize its contribution to the waste bulk.

Neutralized and immobilized reagents are the biggest contribution to the volume of radioactive waste generated by target processing. Consequently the minimization of reagent volume has become an important concern in designing chemistry procedures. Past ion exchange procedures concentrated on using one large column to perform the complete separation and recovery of an isotope. This old approach resulted in the production of large reagent volumes. A significant reduction in reagent waste volumes from ion exchange methods has come from using one ion exchange column to separate

the bulk of target material from radionuclides of interest, then employing a second smaller column or series of smaller columns for the final isolation of the nuclides of interest. This improvement has been especially useful in reducing waste volumes in procedures employed to recover cadmium-109 from indium targets and strontium-82 from molybdenum targets. An added benefit has been improved purity of these radioisotope products.

Other Major Sources of Waste

Materials brought into the hot cell facility are treated as if potentially contaminated due to the difficulty of proving otherwise. All discarded packing materials, laboratory equipment, reagents, instruments, tools, and metal stock became part of the radioactive waste stream. In the past this represented a substantial volume of low level radioactive waste. Unpacking materials outside of the radiologically controlled area coupled with the policy of "bringing in only what you need" has significantly reduced the volume of such waste.

Some material must be unpacked in a radiologically controlled area due to potential contamination of the articles. The greatest contributor to this type of waste had been returned lead shields and packing material from radioisotope product shipments. This source of waste has been virtually eliminated by shipping products in an inexpensive non-returnable shield. When radioisotope shipments require a large returnable lead shield, a reusable high density polyethylene foam box liner minimizes the waste. Formerly a disposable liner was used and resulted in more than a cubic foot of waste when each shield was returned.

Future Goals

Future steps toward waste minimization will require more effort to accomplish smaller

gains. The program is currently investigating compaction mechanisms for hot cell waste. A concern is that high concentrations of radioactive material in waste containers may result in increased personnel exposure hazards. Allowing waste to decay before disposal may help alleviate this problem but takes up space in the limited area of the hot cell facility. An attractive solution to this problem is to recover radioisotopes that are now part of the waste stream and provide them as products. Accordingly, we are developing a procedure to isolate sodium-22 from our target encapsulation and are recovering technetium-95m, arsenic-73, selenium-75, and germanium-68 from molybdenum target solutions.

CONCLUSIONS

Substitution of non-hazardous materials has allowed us to continue to produce many needed radioisotope products without the generation of mixed waste. We have found that chemical separations such as precipitations can work just as well as organic extractants and are easily adapted to hot cell use. Ion exchange and electrochemical procedures can be modified without the use of hazardous organic compounds. Radiation shielding made of materials other than lead usually requires more bulk but often has greater strength and is easier to fabricate. We are successfully using magnetite, brass, copper, tantalum, tungsten, and steel, in place of lead.

Many changes have been made to minimize the volume of waste generated by the program. Modifying chemical separation procedures to minimize reagent waste volumes has substantially reduced the quantity of waste leaving the hot cells. Not introducing materials into the radiologically controlled area until they are needed has considerably reduced our volume of the waste with a minimum of effort. These improvements, coupled with the approach of buying only what is needed and disposing of only the damaged portion of a piece of equipment have not just reduced the volume of waste, but have also provided financial savings. A significant part of our program now involves

waste-minimization research and development. Future in-roads into this problem will be realized as this work matures.

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